



Digital Autoradiography technique an efficient tool for sampling procedure

P. Fichet, R. Haudebourg, C. Mougel

► To cite this version:

P. Fichet, R. Haudebourg, C. Mougel. Digital Autoradiography technique an efficient tool for sampling procedure. DandRS 2016, Jul 2016, Pittsburgh, United States. cea-02438715

HAL Id: cea-02438715

<https://cea.hal.science/cea-02438715>

Submitted on 28 Feb 2020

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

DE LA RECHERCHE À L'INDUSTRIE



www.cea.fr

DIGITAL AUTORADIOGRAPHY TECHNIQUE AN EFFICIENT TOOL FOR SAMPLING PROCEDURE

PASCAL FICHET, RAPHAËL HAUDEBOURG, CAROLINE MOUGEL

DEN – SERVICE D'ETUDES ANALYTIQUES ET DE RÉACTIVITÉ DES SURFACES

(SEARS), CEA, UNIVERSITÉ PARIS-SACLAY, F-91191, GIF SUR YVETTE, FRANCE

MAIL: PASCAL.FICHET@CEA.FR

AUGUST 2ND 2016

Overview

- Context and LASE laboratory : Analytical Support to Facilities Laboratory
- Digital Autoradiography Technique
- Digital Autoradiography in support of sampling processes
- Digital Autoradiography: improvement of the selectivity
- Conclusions

Overview

- Context and LASE laboratory : Analytical Support to Facilities Laboratory
- Digital Autoradiography Technique
- Digital Autoradiography in support of sampling processes
- Digital Autoradiography: improvement of the selectivity
- Conclusions

CONTEXT: MANAGEMENT OF NUCLEAR WASTES

↳ **Who?** The National Radioactive Waste Management Agency **ANDRA** is in charge of the long-term management of radioactive wastes in France

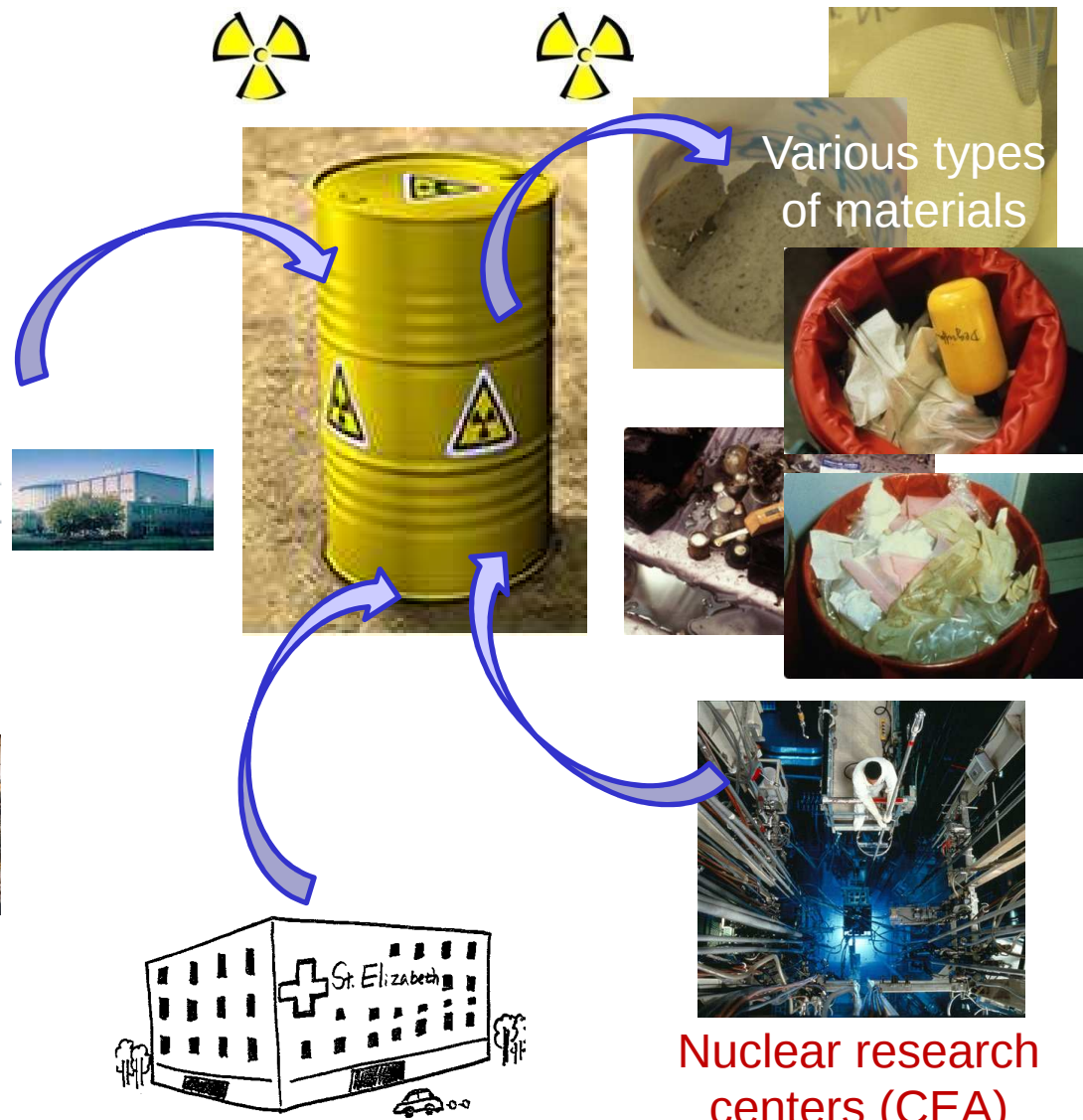


⇒ Classification of radioactive wastes as a function of their management

<i>Activity - Half-life</i>	<i>Very short-half-life < 100 days</i>	<i>Short half-life ≤ 31 years</i>	<i>Long half-life > 31 years</i>
Very low level waste	Management by radioactive decay	Surface disposal facility (CSTFA Aube facility)	
Low level waste		Surface disposal facility (CSFMA Aube facility)	Near-surface disposal facility being studied
Intermediate level waste			Deep disposal facility at 500 meters being studied
High level waste		Deep disposal facility at 500 meters being studied	

CONTEXT: ORIGIN OF LOW AND INTERMEDIATE LEVEL NUCLEAR WASTES

Nuclear power industry (EDF, AREVA)



Nuclear research centers (CEA)

Hospitals

NUCLEAR WASTES ANALYSIS REQUIRED BY AUTHORITIES

👉 **Why? ANDRA** requests a characterization of nuclear wastes and specifies acceptance criteria for packages that waste producers have to respect

Andra Sites Producteurs - Un industriel du cycle électronucléaire (EDF, AREVA, CEA...) - Windows Internet Explorer

http://www.andra.fr/producteurs/pages/fr/menu4/un-industriel-du-cycle-electronucleaire-edf-areva-cea...:1653.html

Fichier Edition Affichage Favoris Outils ?

Liens Arts Programme association Milpat Auchan Billet carrefour Choisir une destination en France - Gîtes de France Cinéma Palaiseau (91120) Cinéma Palaiseau Clubvisioscene

Convertir Sélectionner

Andra Sites Producteurs - Un industriel du cycle élect...

Recherche: OK

L'ANNÉE DES 2011 ANDRA la maîtrise des déchets radioactifs

Producteurs

Accueil

Vous êtes un particulier, un établissement scolaire, une collectivité ou un service de secours

Vous êtes un autre professionnel ou un industriel hors filière électronucléaire

Vous êtes un hôpital, une université, un laboratoire de recherche

Vous êtes un industriel du cycle électronucléaire (EDF, AREVA, CEA...)

Commander des étiquettes codes à barres

Déclarer vos matières et déchets radioactifs pour l'inventaire national

Vous êtes un industriel du cycle électronucléaire (EDF, AREVA, CEA)

L'Andra est à votre disposition pour vous assister et vous conseiller dans votre recherche de filière d'élimination et pour définir avec vous les modalités de caractérisation, tri, traitement et conditionnement de vos déchets.

Après agrément et acceptation des colis de déchets radioactifs par l'Andra (autorisation de livraison), ceux-ci sont directement acheminés vers le CSTFA (Centre de stockage des déchets de très faible activité) ou le CSFMA (Centre de stockage de faible et moyenne activité) de l'Andra.

Afin d'assurer le suivi de vos productions de colis de déchets radioactifs et de faire le point sur vos "problématiques déchets", des rendez-vous réguliers sont organisés, à votre initiative ou à celle de l'Andra.

En tant qu'industriel du cycle électronucléaire, après vous être identifié, nous vous invitons à prendre connaissance des spécifications correspondantes à chaque site de stockage de l'Andra.

Partager

Déchargement des colis de déchets au terminal ferroviaire de Brienne-le-Château à destination du CSFMA dans l'Aube.

La majorité des déchets produits par les industriels et les laboratoires du cycle électronucléaire sont de très faible activité (TFA) ou de faible et moyenne activité à vie courte (FMA-VC).

Identifiant:

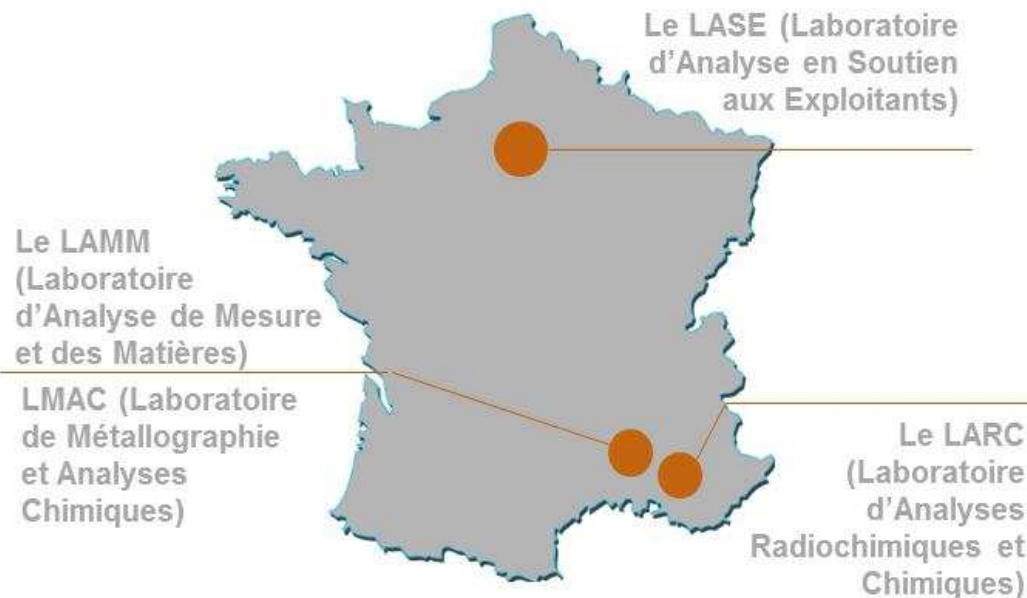
Mot de passe:

Valider

👉 **Why? Characterization is one of the essential step in decommissioning projects**

ANALYTICAL CAPABILITY OF NETLAB

NetLab associated to **NetLab** = A CEA network of analytics and experimental tools (laboratories located on CEA sites) to assist nuclear operators during characterization programs associated to dismantling process.



High Level

Intermediate level

Low level



LASE LABORATORY

(25 km at the south of Paris)



- ❑ Different characterization techniques for low and intermediate level wastes.
 - ❑ Destructive analysis (sample = 1 g)
 - ❑ Radiochemistry
 - ❑ Alpha, Gamma, LSC
 - ❑ Elemental analysis
- ❖ In situ technique: **Autoradiography**



LASE: ANALYTICAL SUPPORT TO FACILITIES LABORATORY FRANCE

- ANALYSES ARE REQUIRED TO ASSESS THE SAFETY OF THE REPOSITORY SITES.
- LOTS OF MATRICES TO BE CONSIDERED, **Validation required**

Elemental analysis



23 fume cupboards
Radiochemistry



Nuclear analyses

CHARACTERIZATION OF LOW AND INTERMEDIATE LEVEL NUCLEAR WASTES

⇒ Mission of Analytical support to facilities Laboratory at CEA-Saclay

ANALYSIS OF ELEMENTS (RADIOACTIVE OR NOT) PRESENT AT TRACE LEVEL IN VARIOUS MATRICES

Chemical and radiological
characterization
of radioactive materials

Toxic elements, organic ligands, TOC,
anions, cations

Wipes, technological wastes (tissues,
gloves), concretes, ion exchange resins
embedded in organic polymers, metals,
muds, sludges, oils...

**Radionuclides determined after
radiochemistry ($A \sim 0.1 \text{Bq.g}^{-1}$)**

^3H , ^{14}C , ^{36}Cl , ^{55}Fe , ^{59}Ni , ^{63}Ni , ^{90}Sr , ^{93}Mo , ^{93}Zr ,
 $^{93\text{m}}\text{Nb}$, ^{94}Nb , $^{108\text{m}}\text{Ag}$, $^{121\text{m}}\text{Sn}$, ^{129}I , ^{151}Sm , ^{241}Pu ,
 238 et ^{240}Pu , ^{239}Pu , ^{241}Am , ^{243}Am , ^{232}U , ^{234}U ,
 ^{235}U , ^{238}U



⇒ Main Radionuclides that must be investigated in priority

❖ Easy to measure:

❖ gamma emitters

❖ Difficult to measure

❖ Alpha emitters

❖ Beta emitters

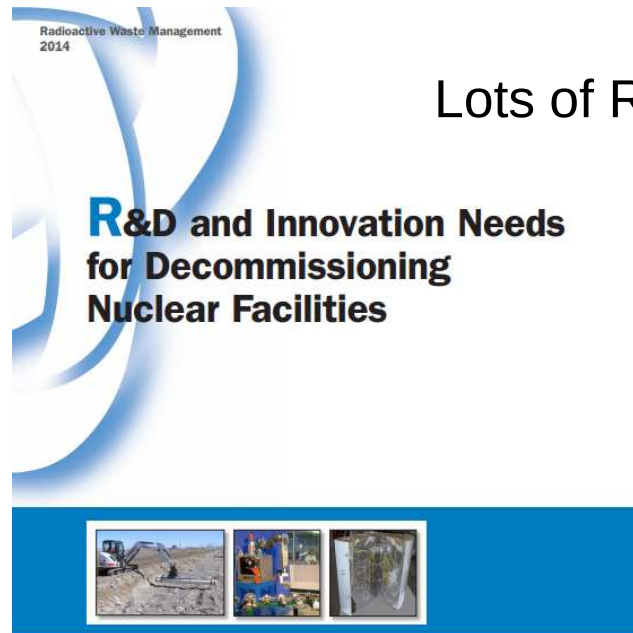
- H-3
- C-14
- Cl-36
- Sr-90

CHARACTERIZATION IS VERY IMPORTANT FOR D&D PROJECTS

Destructive techniques need less than 1 g of sample for digestion process



Lots of Radwastes must be characterized



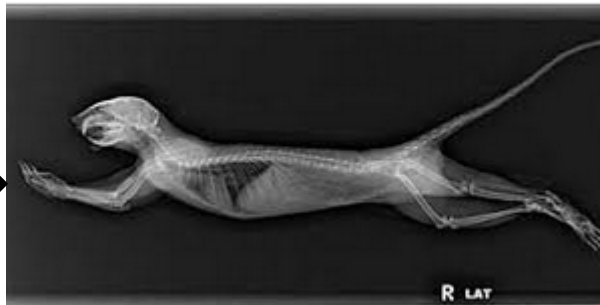
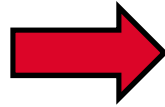
Innovation is required for in situ techniques, for techniques allowing a better sampling process

Overview

- Context and LASE laboratory : Analytical Support to Facilities Laboratory
- Digital Autoradiography Technique
- Digital Autoradiography in support of sampling processes
- Digital Autoradiography: improvement of the selectivity
- Conclusions

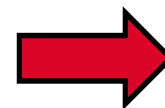
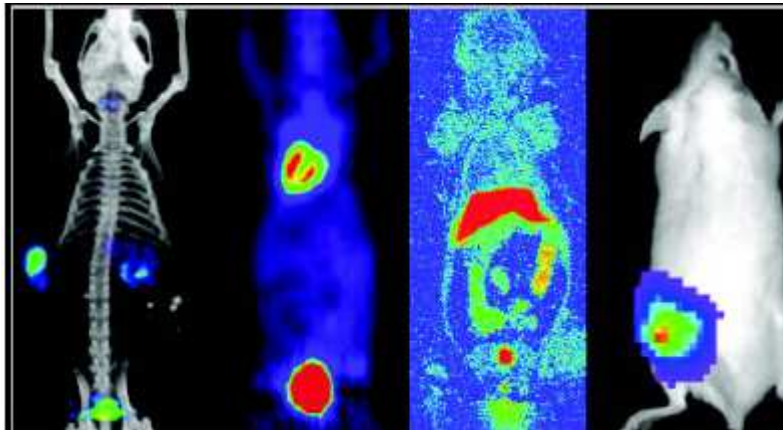
Radiography

Source



Detector

Autoradiography

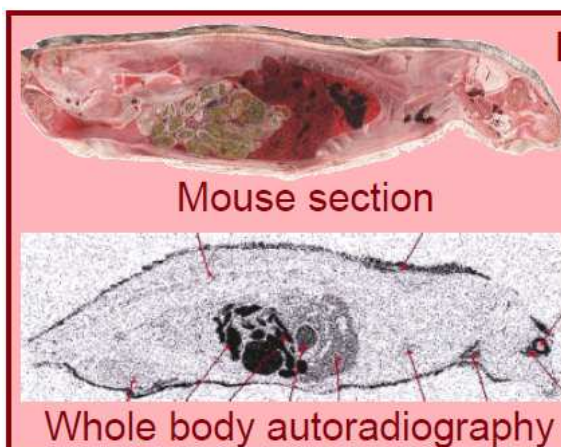
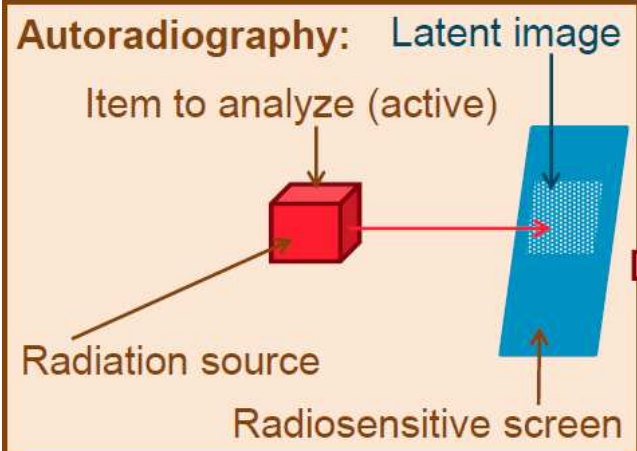


Detector

Interests of biologists: H, C, S, ... radionuclides
difficult to measure

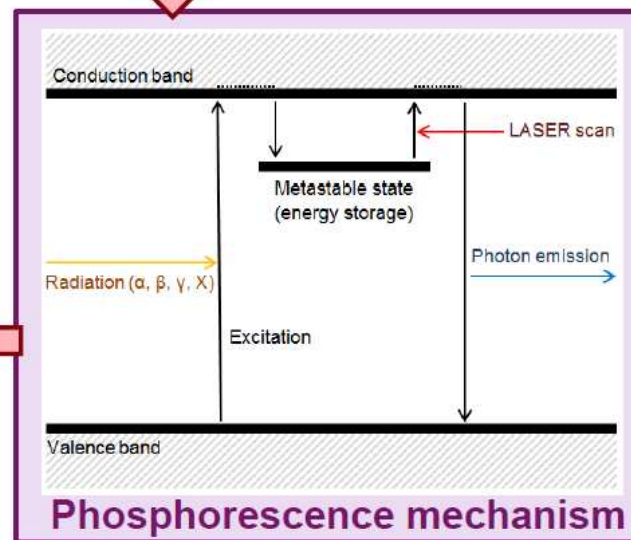


Digital Autoradiography



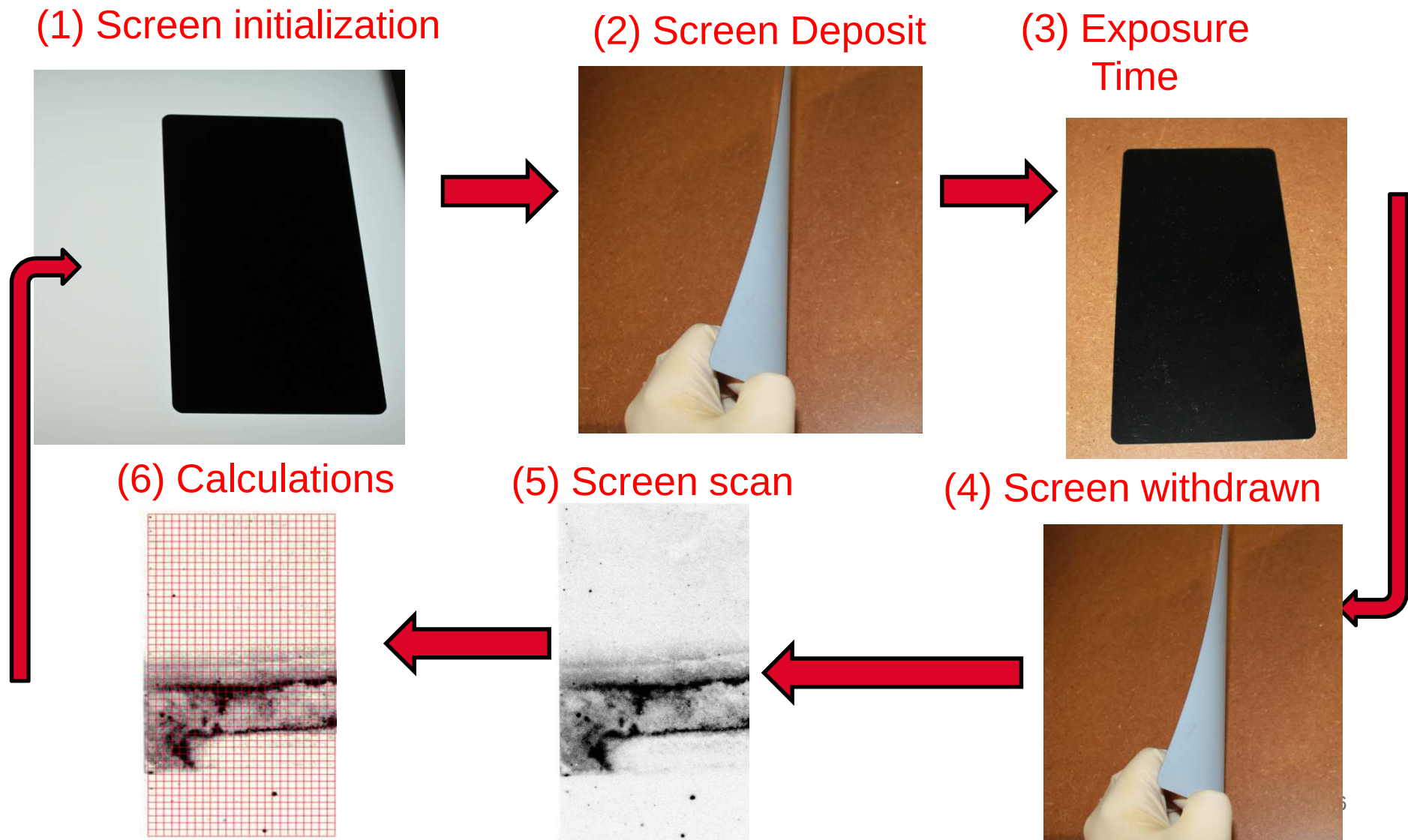
Initial application:
Biomolecular
research

Consequences:
Small screens
High resolution
Low sensitivity

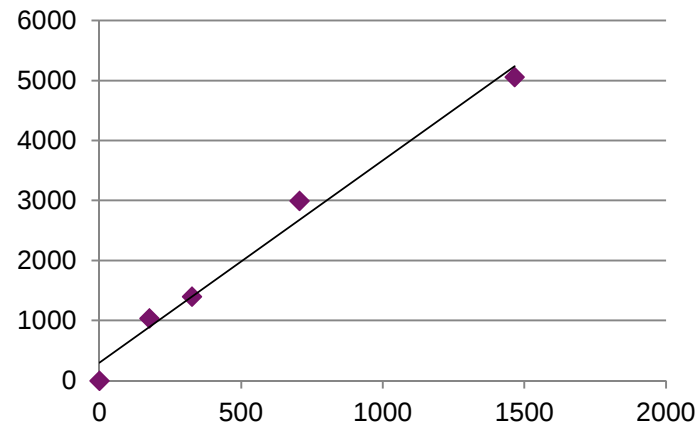


**Image of radioactivity = efficient tool for sampling
procedure**

Screens can be reused hundreds of times

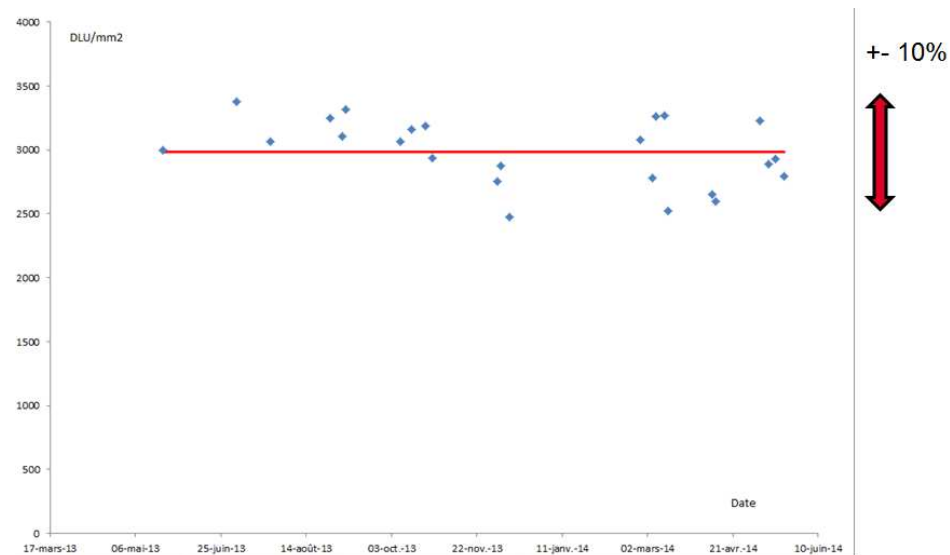


- Semi quantitative values are achievable.



- Repeatability corresponds only to a few percent

Same H-3
analysis



Overview

- Context and LASE laboratory : Analytical Support to Facilities Laboratory
- Digital Autoradiography Technique
- Digital Autoradiography in support of sampling processes
- Digital Autoradiography: improvement of the selectivity
- Conclusions

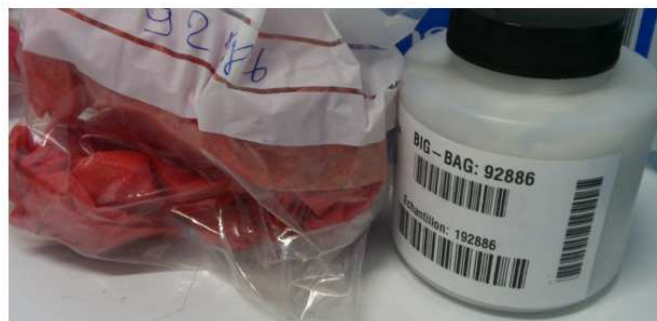
SAMPLING PROBLEMS FOR RADIOCHEMICAL ANALYSIS



Caisse 95430



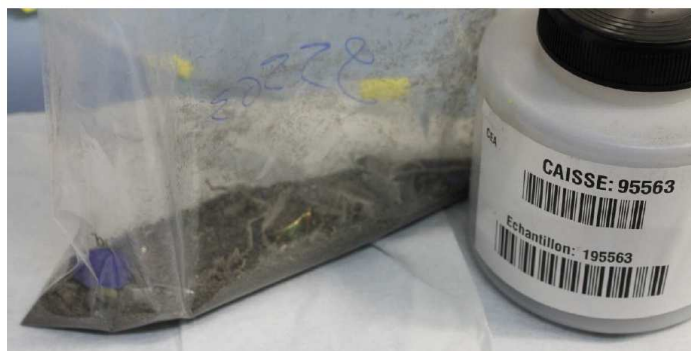
Echantillon 92886



Caisse 95434



Echantillon 95563



SAC 2 95485



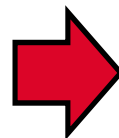
How to
collect 1g or
even less?

DIFFERENT PROCEDURES FOR MEASUREMENTS



In Situ Sampling

Classical sampling process by **wipe tests** = problems for non labile radioactivity



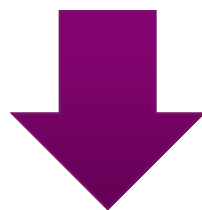
Digestion or leaching
Alpha spectrometers
Liquid Scintillation Counting
Gamma measurements

Laboratory measurements



Container

Autoradiography



In situ measurements by Autoradiography

EXAMPLE OF HOME MADE STANDARDS

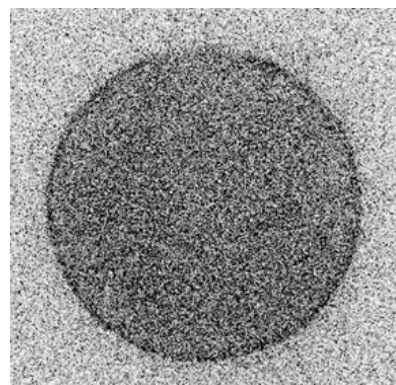
Preparation of homemade standards for beta analysis: CONCRETES

Concrete spiked at the beginning of the preparation of the cement

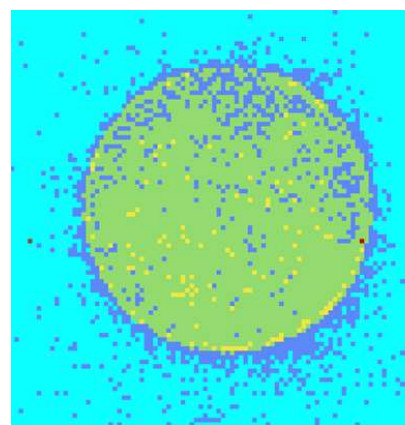
Can be spiked with numerous radionuclides H-3, C-14, Gamma emitters...)



Homogeneity on concrete surface (here H-3 and C-14)



Raw signal



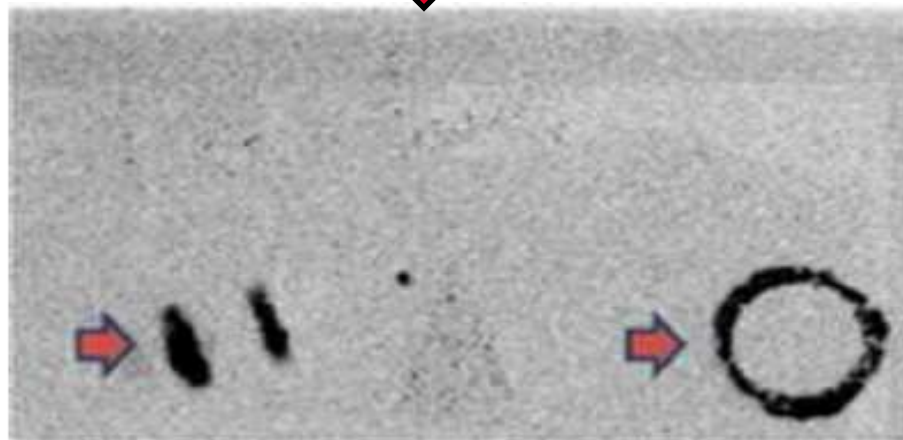
Validated Data DLU
2306.30
1890.22
1474.14
1058.06
641.98
225.90

Numerical treatment to evaluate the homogeneity by digital autoradiography

EXAMPLE OF TRITIATED WASTES

Sampling process for **tritiated** wastes

Autoradiography



Destructive measurement of H-3 is done by pyrolysis followed by Liquid Scintillation Counting (LSC)



Sample = 1 g

H-3 AND C-14 ANALYSIS ON WASTES

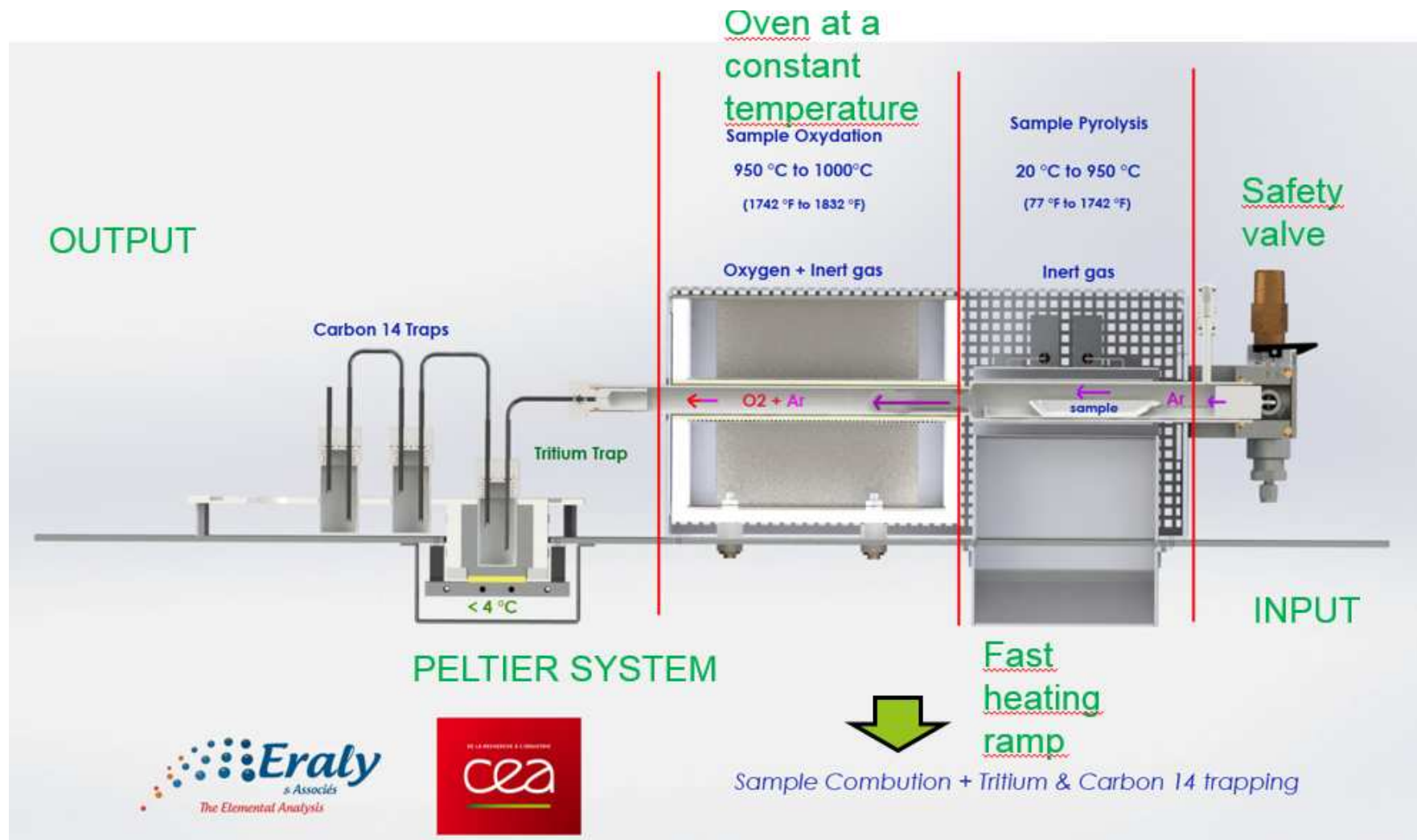
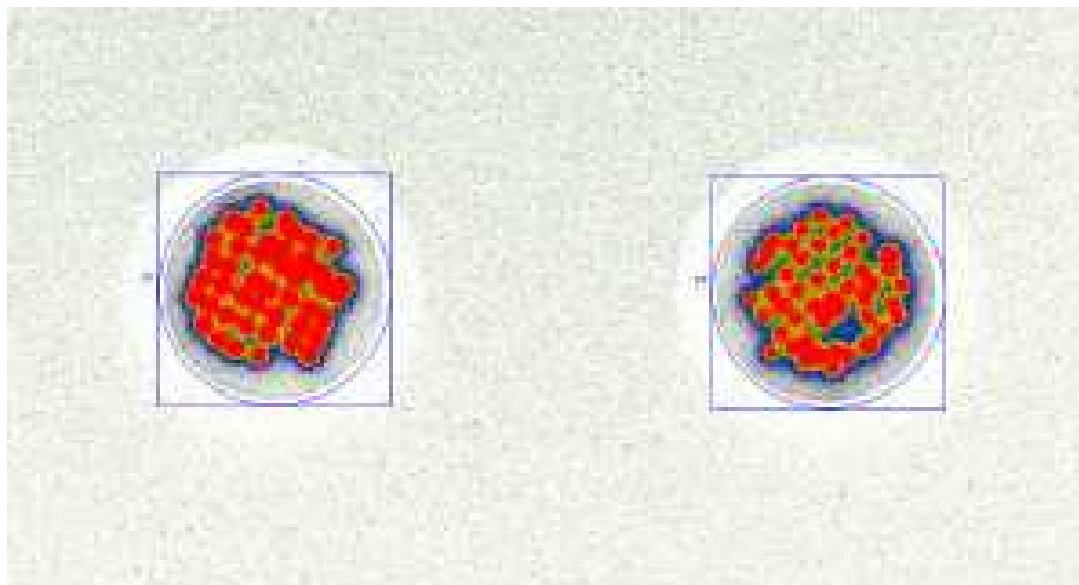


IMAGE OBTAINED WITH HIGH RESOLUTION



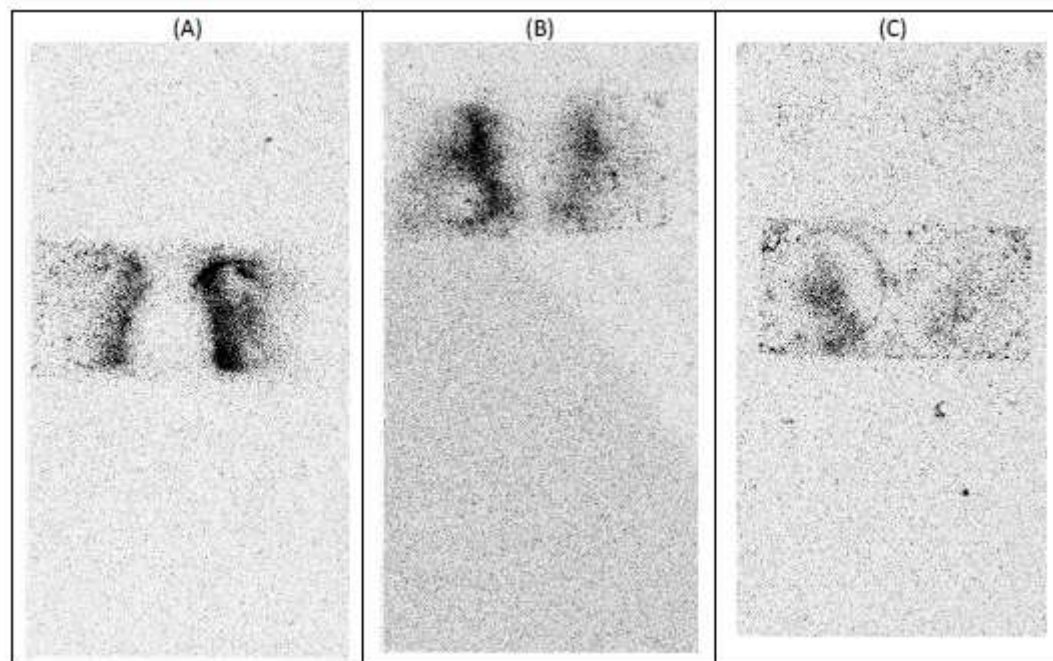
Cl-36

Sr-90

Study of sealed source
by digital
autoradiography –
resolution used = 40 μm

EXAMPLE OF WASTES CONTAINING URANIUM

Only 3 wipes among 7
contained Uranium.



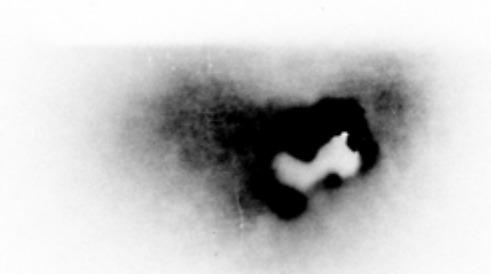
After studies with a destructive method: wipe digestion followed by alpha spectrometry and/or ICP-MS, detection limit was determined at **0.2 Bq/wipe** for Uranium (more sensitive than alpha spectroscopy).

HOW TO COLLECT SAMPLES ON CORES

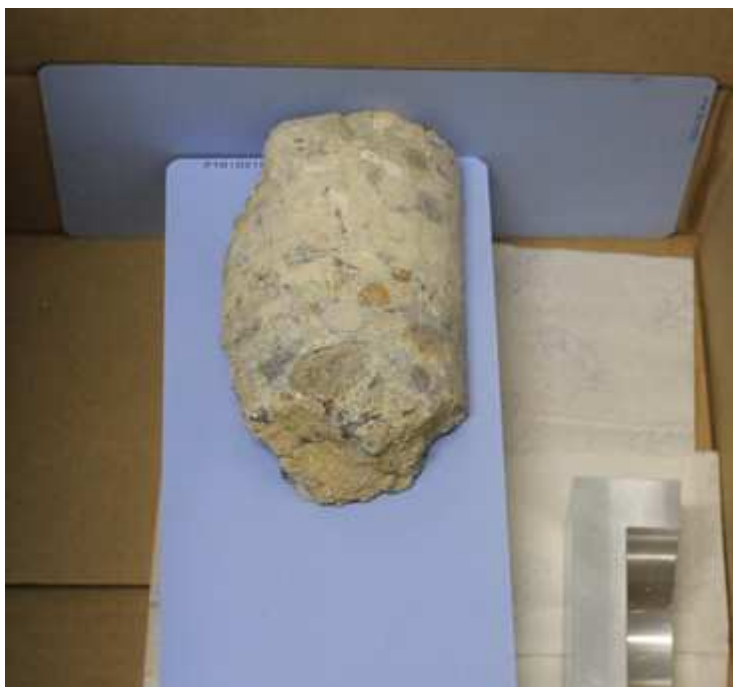
After surface analysis on D&D sites, determination of 3D contamination



Core made of
concrete
containing C-14



Activity on surface



Interesting
sampling



Activity
in depth

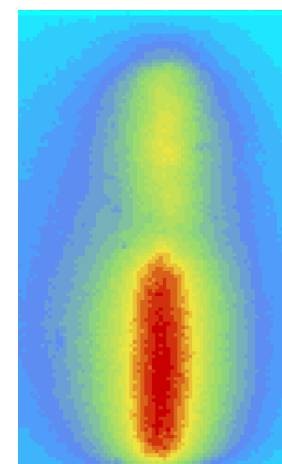
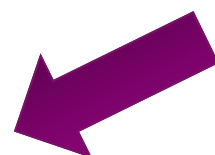
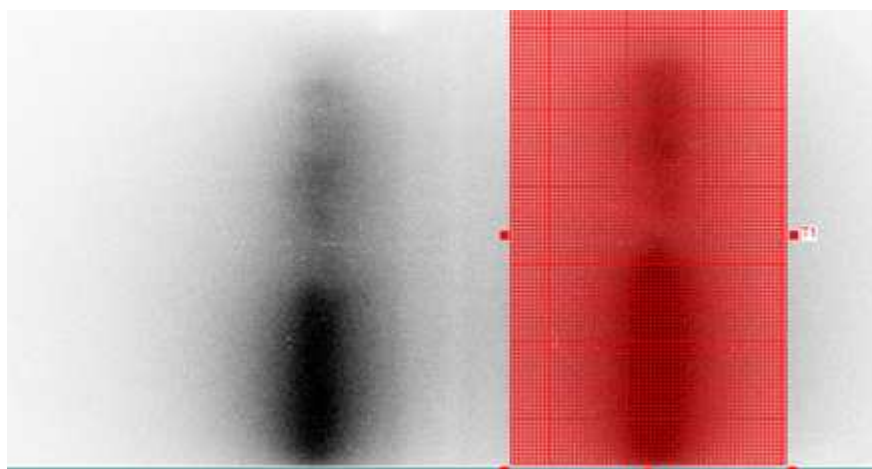
Image in real
scale

HOW TO COLLECT SAMPLES ON CORES WITH HIGH AMOUNT OF RADIOACTIVITY ?

Study of alumina beads containing I-129 located inside a plastic tube



Plastic deposited on a film for **15 minutes**

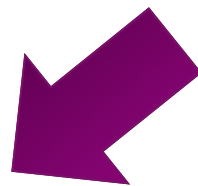
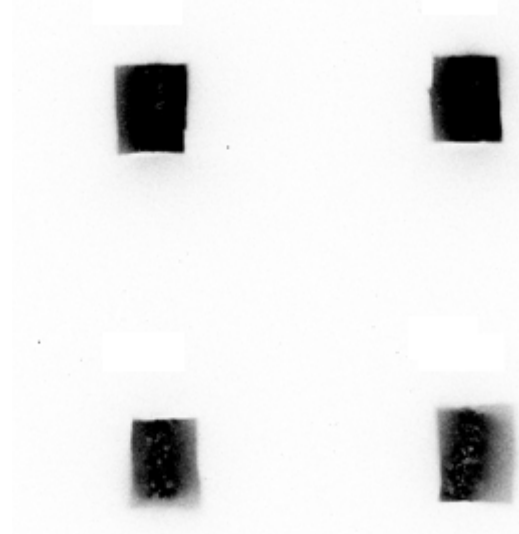
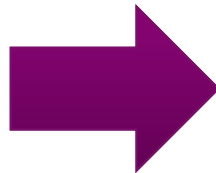


DA as efficient solution for sampling process

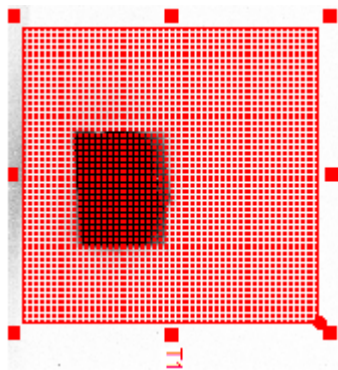
SAMPLING PROBLEM ON GRAPHITES

4 small cores on the same screen

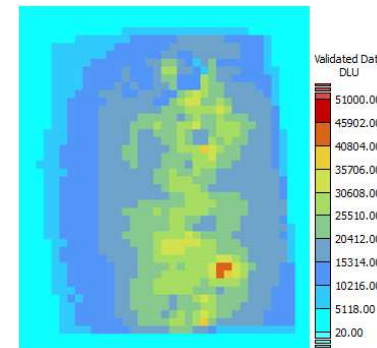
Graphite cores mainly containing C-14



Calculations



Homogeneity of the contamination at a very low scale



Digital Autoradiography is a non destructive method to assess the homogeneity

SAMPLING PROBLEM ON STEEL MATERIALS



Massive steel
samples
investigated by
digital
autoradiography

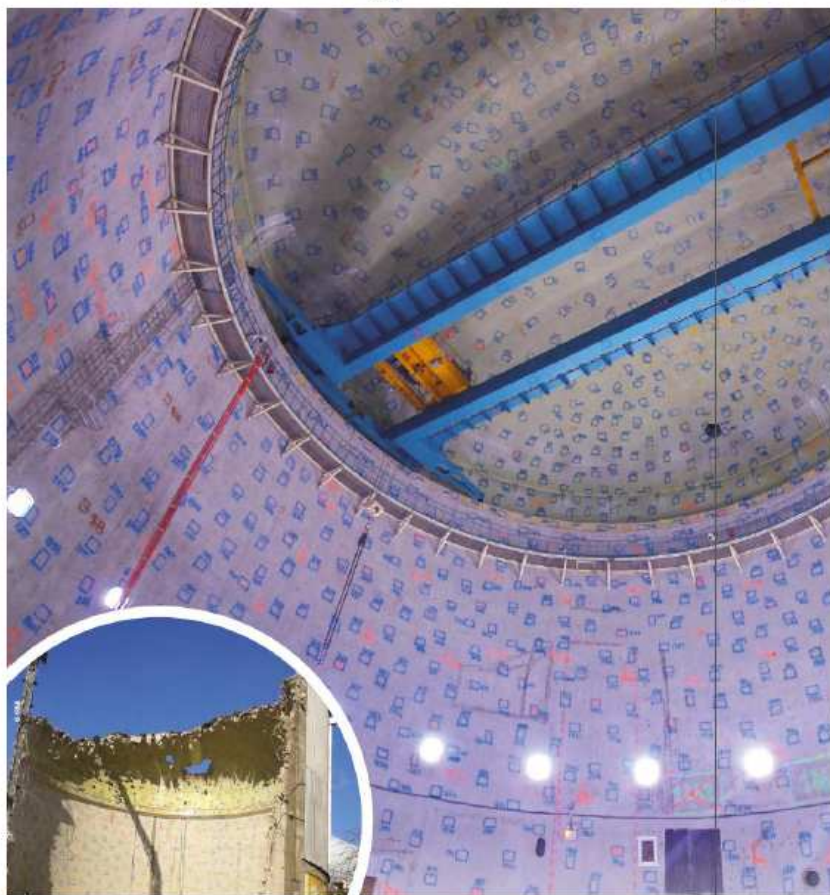
For some
samples only one
side is
contaminated

Contamination location and homogeneity

SAMPLING FOR FINAL CONTROL TO ASSESS THE CLEARANCE LEVEL

Decommissioning requires characterization at very low level. Requirement of in situ technique to improve sampling process. 1 g sample needed for destructive analysis.

Decommissioning and dismantling

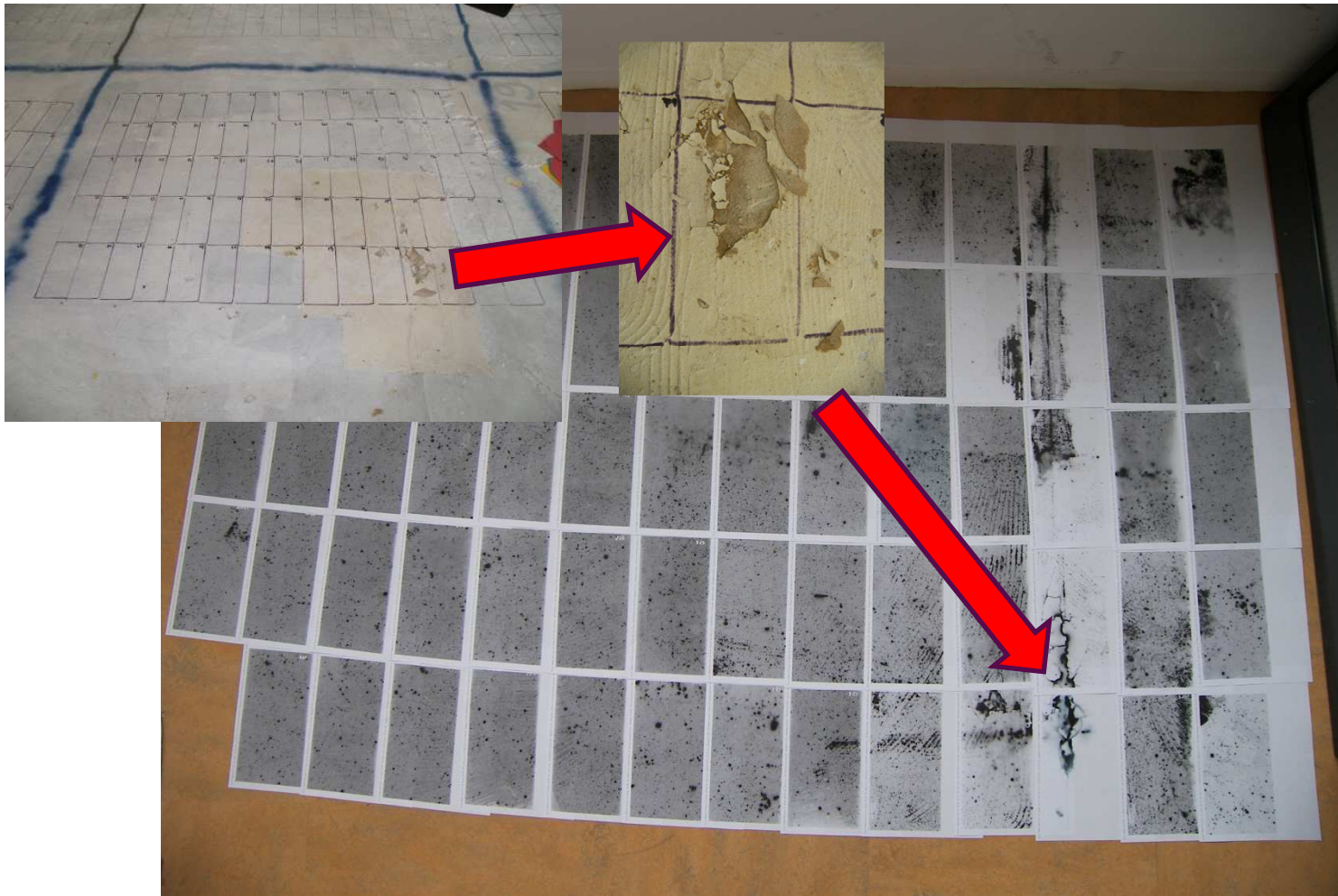


Mélusine, Grenoble



Sampling process on floor

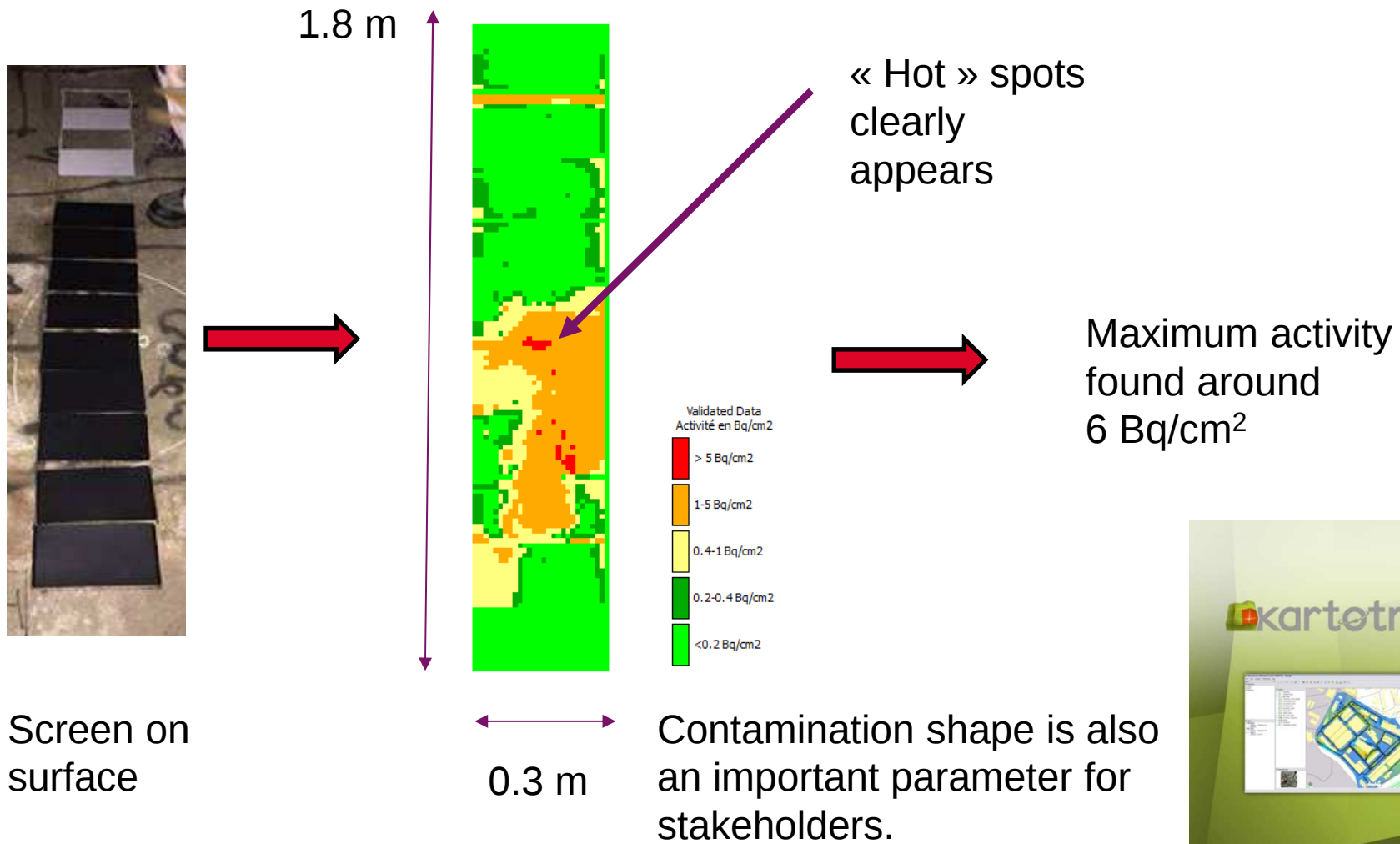
A grid corresponding to 70 screens = 5 m²



Radioactivity
image, here
C-14
Localisation
of the sample
for destructive
analysis is
clear

Autoradiography with raw images

OTHER EXAMPLE FOR URANIUM MEASUREMENTS



OTHER EXAMPLE FOR TRITIUM SAMPLING

Current development: geostatistical approach



Current investigation rate:
2 weeks / 100 m²

Conclusion:

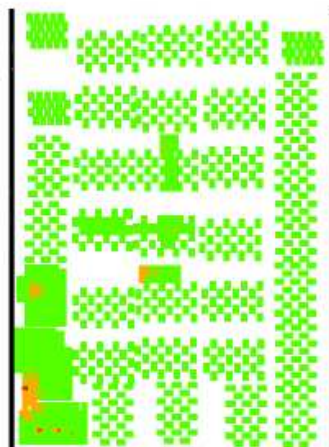
Contamination accurate location

Representative and limited sampling enabling

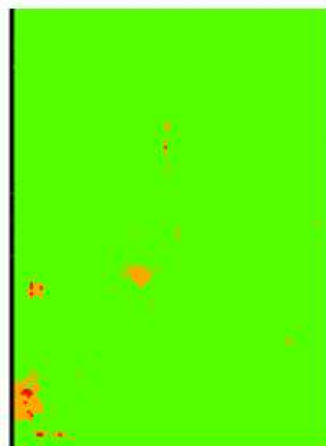


29	28	15	14	1
30	27	16	13	2
31	26	17	12	3
32	25	18	11	4
33	24	19	10	5
34	23	20	9	6
35	22	21	8	7

Zoning

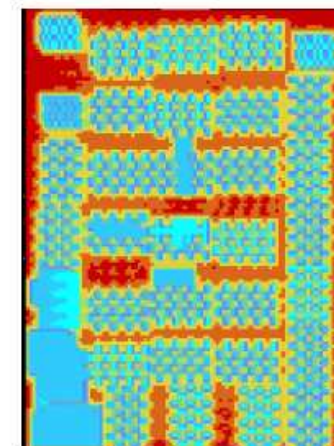


Effective mapping



Geostatistical mapping

+



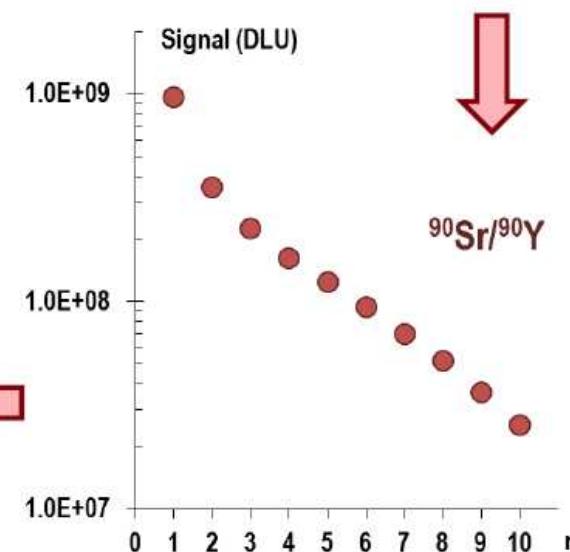
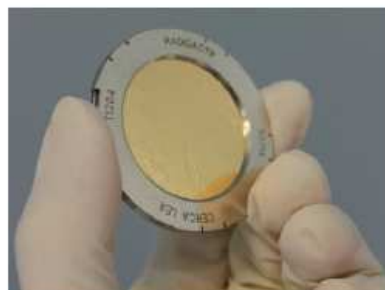
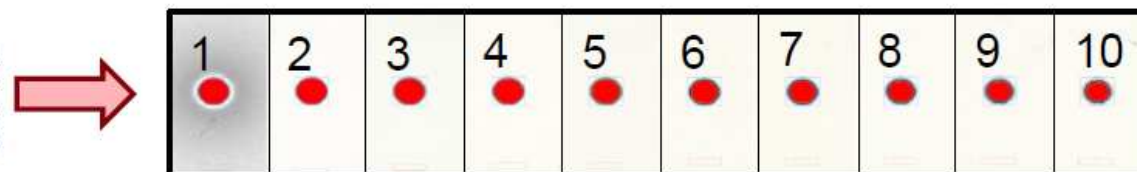
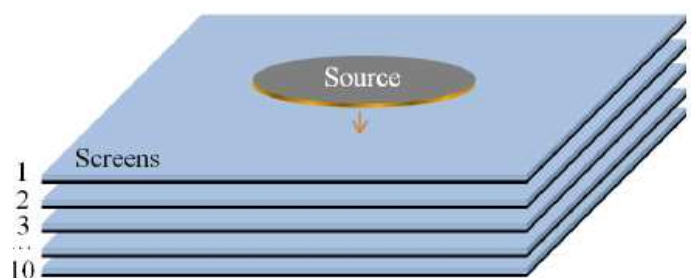
Variance plot

Overview

- Context and LASE laboratory : Analytical Support to Facilities Laboratory
- Digital Autoradiography Technique
- Digital Autoradiography in support of sampling processes
- **Digital Autoradiography: improvement of the selectivity**
- Conclusions

IMPROVEMENT OF SELECTIVITY

Screen stacking method (1/2)



Intermediate precision

example of $^{90}\text{Sr}/^{90}\text{Y}$:

2 point sealed sources from LASE

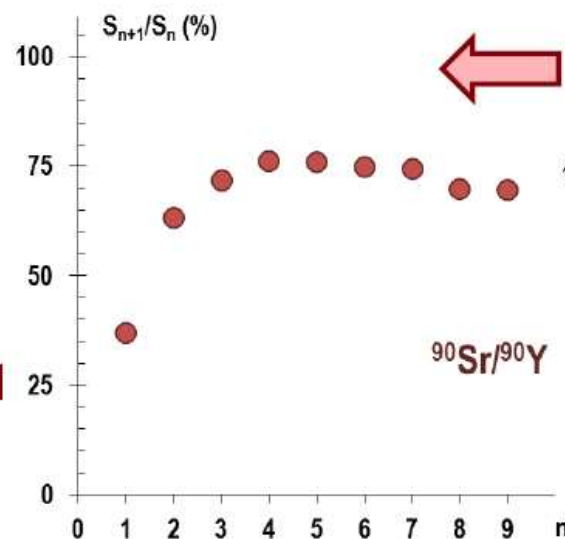
5 sealed filtrates from LASE

1 point sealed source from INSTN

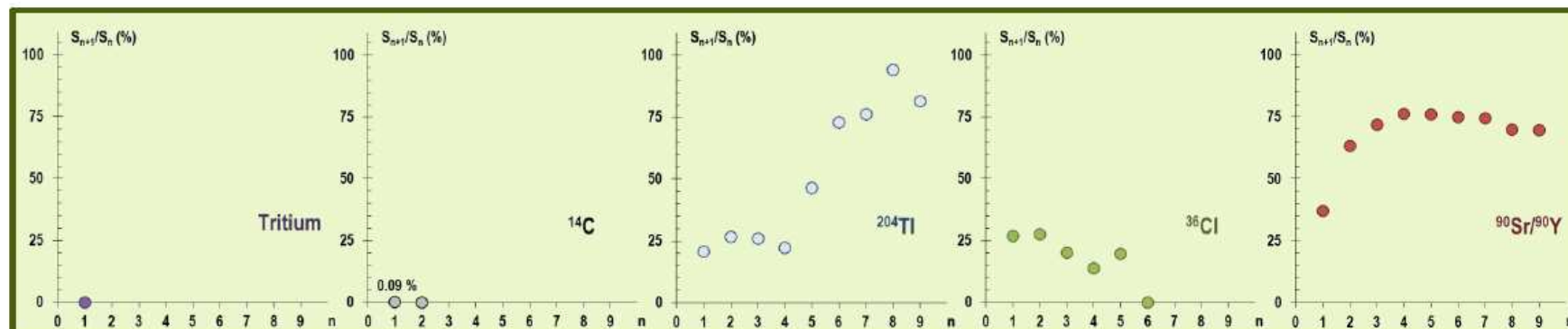
2 area sources from INSTN

$$S_2/S_1 = 33 \pm 6 \%$$

$$S_3/S_2 = 59 \pm 6 \%$$



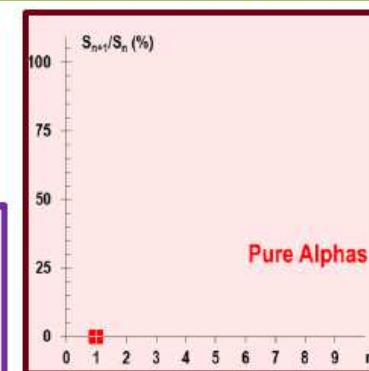
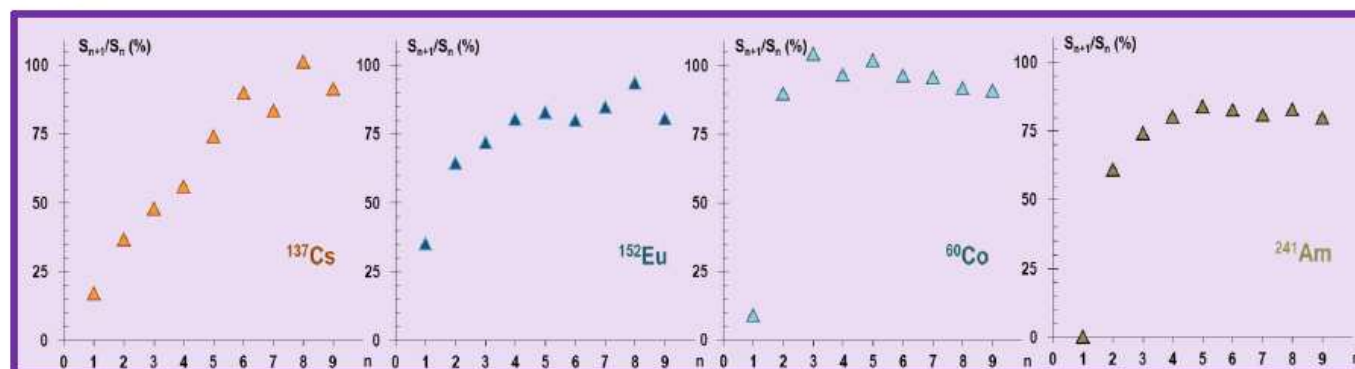
Screen stacking method (2/2)



β

Radionuclides "signatures"

$\beta+\gamma$, $\alpha+X$



α

Overview

- Context and LASE laboratory : Analytical Support to Facilities Laboratory
- Digital Autoradiography Technique
- Digital Autoradiography in support of sampling processes
- Digital Autoradiography: improvement of the selectivity
- **Conclusions**

- ❑ Digital Autoradiography was used first because the technique is sensitive to difficult to measure radionuclides.
- ❑ Autoradiography appears to be a very interesting technique for sampling processes.
- ❑ Different developments are currently in R&D for mapping applications, to improve the sensitivity, to try to find possibilities to improve the selectivity.
- ❑ The LASE laboratory participates in numerous intercomparison tests where no sampling problem is usually encountered however it is totally different for radiochemical analysis required for radwastes matrices.

References:

- 1) «Tritium analysis in building dismantling process using digital autoradiography » P Fichet, F Bresson, A Leskinen, F Goutelard, J Ikonen, M Siitari-Kauppi, Journal of Radioanalytical and Nuclear Chemistry, 291: 869-875 (2012).
- 2) « Digital autoradiography (DA) in quantification of trace level beta emitters on concrete » A Leskinen, P Fichet, M Siitari-Kauppi, F Goutelard, Journal of Radioanalytical and Nuclear Chemistry 298 : 153-161 (2013).
- 3) « A non-destructive and on-site digital autoradiography-based tool to identify contaminating radionuclide in nuclear wastes and facilities to be dismantled» R Haudebourg, P Fichet : J Radioanal Nucl Chem to be published 2016

Oral presentations:

- 1) «Radiological mapping of facilities under dismantling process by Digital Autoradiography», Waste Management Phoenix 2015.
- 2) «Digital Autoradiography : a novel complementary technique for the investigation of radioactive contamination in nuclear facilities under dismantlement» ANNIMA 2015, Lisbon, R Haudebourg, P Fichet, F Goutelard.
- 3) « Radiological mapping of nuclear facilities under dismantlement drawn up by Digital Autoradiography technique» R. Haudebourg, P. Fichet, F. Goutelard, Global 2015 Paris

Thank you for your attention



Commissariat à l'énergie atomique et aux énergies alternatives
Centre de Saclay | 91191 Gif-sur-Yvette Cedex
T. +33 (0)1 69 08 26 27 | F. +33 (0)1 69 08 43 23

Etablissement public à caractère industriel et commercial | RCS Paris B 775 685 019